

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
6 April 2006 (06.04.2006)

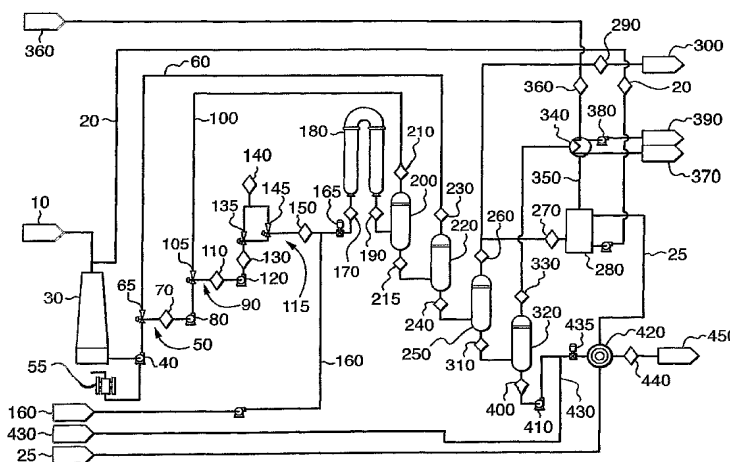
PCT

(10) International Publication Number
WO 2006/034590 A1

- (51) International Patent Classification⁷: **B09B 3/00**, C12P 7/10, 19/02, 19/14
- (21) International Application Number: PCT/CA2005/001493
- (22) International Filing Date: 30 September 2005 (30.09.2005)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data: 60/614,700 30 September 2004 (30.09.2004) US
- (71) Applicant (for all designated States except US): **IOGEN ENERGY CORPORATION** [CA/CA]; 8 Colonnade Road, Ottawa, Ontario K2E 7M6 (CA).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): **FOODY, Brian** [CA/CA]; 1 Linden Terrace, Ottawa, Ontario K1S 1Z1 (CA). **ANAND, Vijay** [CA/CA]; 925 Renoir Street, Brossard, Quebec J4X 2H1 (CA). **REA, David** [CA/CA]; 5363 McLean Crescent, Manotick, Ontario K4M 1E3 (CA).
- (74) Agents: **SECHLEY, Konrad, A.** et al.; Gowling Lafleur Henderson LLP, 160 Elgin Street, suite 2600, Ottawa, Ontario K1P 1C3 (CA).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, LY, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SM, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IS, IT, LT, LU, LV, MC, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).
- Published:
— with international search report
— with amended claims

[Continued on next page]

(54) Title: CONTINUOUS FLOWING PRE-TREATMENT SYSTEM WITH STEAM RECOVERY



(57) Abstract: The present invention is directed to a continuous process for pretreating a lignocellulosic feedstock. A feedstock slurry is provided, which has a solids concentration of 10% to 30% by weight. The feedstock slurry is pumped through a heating train of at least two stages, each stage including a pump to increase stage pressure and a direct steam injection to heat the feedstock slurry. Acid is also added to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train; the acid is added at a concentration of 0.2% to 12% weight of acid on weight of initial feedstock. A heated, acidified feedstock slurry is thus produced. The heated, acidified feedstock slurry is flowed through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes. After the slurry exits the pretreatment reactor, it is cooled using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing. The steam energy, at or near its flash pressure, from two or more than two of the flashing stages is used to heat the feedstock slurry in the heating train.



For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

CONTINUOUS FLOWING PRE-TREATMENT SYSTEM WITH STEAM RECOVERY

[0001] The present invention relates to a pre-treatment process in the conversion of lignocellulosic feedstocks into sugars. More specifically, the present invention relates to a continuously flowing pre-treatment process with simultaneous steam recovery, for the conversion of lignocellulosic feedstocks into sugars.

BACKGROUND OF THE INVENTION

[0002] Fuel ethanol is currently produced from feedstocks such as cornstarch, sugar cane, and sugar beets. However, the production of ethanol from these sources cannot expand much further due to limited farmland suitable for the production of such crops and competing interests with the human and animal food chain. Finally, the use of fossil fuels, with the associated release of carbon dioxide and other products, in the conversion process is a negative environmental impact of the use of these feedstocks.

[0003] The possibility of producing ethanol from cellulose-containing feedstocks such as agricultural wastes, grasses, and forestry wastes has received much attention due to the availability of large amounts of these inexpensive feedstocks, the desirability to avoid burning or landfilling cellulosic waste materials, and the cleanliness of ethanol as a fuel compared to gasoline. In addition, a byproduct of the cellulose conversion process, lignin, can be used as a fuel to power the cellulose conversion process, thereby avoiding the use of fossil fuels. Studies have shown that, taking the entire cycle into account, the use of ethanol produced from cellulose generates close to nil greenhouse gases.

[0004] The cellulosic feedstocks that may be used for ethanol production include (1) agricultural wastes such as stover, including corn stover and soybean stover, corn cobs, rice straw, rice hulls, corn fiber, wheat straw, barley straw, canola straw, oat straw and oat hulls; (2) grasses such as switch grass, miscanthus, cord grass, rye grass and reed canary grass; (3) forestry biomass such as recycled wood pulp fiber, softwood, hardwood and sawdust; and (4) sugar processing residues such as bagasse and beet pulp.

[0005] Cellulose consists of a crystalline structure that is very resistant to breakdown, as is hemicellulose, the second most prevalent component. The conversion of cellulosic fibers to ethanol requires: 1) liberating cellulose and hemicellulose from lignin or increasing the accessibility of cellulose and hemicellulose within the cellulosic feedstock to cellulase enzymes; 2) depolymerizing hemicellulose and cellulose carbohydrate polymers to free sugars; and 3) fermenting the mixed hexose and pentose sugars to ethanol.

[0006] Among well-known methods used to convert cellulose to sugars is an acid hydrolysis process involving the use of steam and acid at a temperature, acid concentration and length of time sufficient to hydrolyze the cellulose to glucose (Grethlein, 1978, J. Appl. Chem. Biotechnol. 28:296-308). The glucose product is then fermented to ethanol using yeast, and the ethanol is recovered and purified by distillation.

[0007] An alternative method of cellulose hydrolysis is an acid prehydrolysis (or pre-treatment) followed by enzymatic hydrolysis. In this sequence, the cellulosic material is first pre-treated using the acid hydrolysis process described above, but at milder temperatures, acid concentration and treatment time. This pre-treatment process is thought to increase the accessibility of cellulose within the cellulosic fibers for subsequent enzymatic conversion steps, but results in little conversion of the cellulose to glucose itself. In the next step, the pre-treated feedstock is adjusted to an appropriate temperature and pH, then submitted to enzymatic conversion by cellulase enzymes.

[0008] The hydrolysis of the cellulose, whether by acid or by cellulase enzymes, is followed by the fermentation of the sugar to ethanol, which is then recovered by distillation.

[0009] The temperatures typically used for acid hydrolysis or pre-treatment correspond to saturated steam pressures of 160 psig to 665 psig. The addition of sulfuric acid improves the reaction of the cellulose and hemicellulose and can shorten the time for pre-treatment from 5-30 minutes to 0.1-5 minutes. Achieving and maintaining these conditions requires a highly pressurized, acid-resistant system. U.S. Patent No. 4,461,648 (Foody) describes equipment and conditions used in steam explosion pre-treatment, in which the feedstock, steam, and sulfuric acid are added to a reaction vessel, known as a steam gun. In the steam gun, steam is added and the steam pressure is increased rapidly to the desired pressure, held at this pressure for a period

of time, followed by sudden explosive decompression. Steam explosion with sulfuric acid added has been the standard pre-treatment process for two decades. It produces pre-treated material that is uniform, has most of the hemicellulose hydrolyzed to simple sugar, and less cellulase enzyme is subsequently required to hydrolyze the cellulose than other pre-treatment processes.

[0010] Although steam explosion and other batch pre-treatment processes prepare feedstocks with highly accessible cellulose, they have inherently low throughput. This requires the use of many steam pre-treatment reactors, which is costly and difficult to operate. Furthermore, it is difficult in batch processes – and in steam explosion in particular – to recover and re-use the steam. The steam requirement to achieve and maintain the feedstock at acid hydrolysis or pre-treatment conditions is high. The loss of this steam represents a significant cost in the ethanol production process.

[0011] The development of a continuous acid pre-treatment process delivering the high degree of accessibility of the cellulose from a steam explosion process, and that can economically recover and re-use steam would be desirable. Some investigators have disclosed flashing of the high pressure steam from the pre-treatment reactor to low pressure and capturing the low pressure steam. In some cases, this has involved the use of a single flashing of steam to low pressure. The low pressure steam is then used to provide a portion of the steam required to pre-heat the slurry prior to pre-treatment. Unfortunately, this recycling of the steam is inefficient and only provides a limited portion of the steam required for pre-treatment. An alternative approach, which involves compressing the flashed steam and then re-using it at a higher pressure, incurs the equipment and operating cost of the compression. Furthermore, in a continuous pre-treatment process it is desirable that the feedstock can be pumped. However, many of the known pre-treatment processes do not make use of pumping and the feedstock which is utilized has a solids content and/or a particle size that is not suitable for pumping. The requirement remains for a continuous pre-treatment system with efficient recovery and re-use of steam and that carries out pumping of the feedstock.

[0012] U.S. Patent No. 4,136,207 (Bender) teaches steam treatment to produce a ruminant feed. The feedstock is saturated with moisture and compacted at 2000 psi to remove air and improve the subsequent penetration of steam. A rotating helical feed screw conveys the

compacted feedstock into a digester. Steam produced by a steam generator is fed into the digester at 200-310 psi. The feedstock proceeds through the digester, at the end of which is a valve to allow volatile byproducts to be withdrawn to a byproduct recovery column, and a product valve for treated solids to exit. Steam, which is collected in a byproduct recovery column, is returned to the digester. However, the low moisture content of the feedstock does not allow for it to be pumped. Furthermore, the recovery of steam by Bender subsequent to steam treatment in the digester is limited to a single flash at low pressure, which is inefficient. The recovered steam is compressed prior to re-use, which itself consumes energy and adds to the cost of the process. The process also does not provide for the use of acid, which is required for rapid reaction and the production of maximum accessibility of the cellulose in an enzymatic conversion process.

[0013] U.S. Patent No. 5,366,558 (Brink) describes a continuous acid hydrolysis process that occurs in several stages. The first stage is a steam treatment in the absence of acid. The material is then mechanically disintegrated to a very small particle size, acidified, and sensitized with oxygen. The sensitized material is then heated with steam for the final hydrolysis reaction. The material is washed countercurrently, with a sugar stream and lignin being the products. As multiple reaction steps at high temperature and pressure are required, the risk of severe degradation of the feedstock increases. This in turn limits the level of cellulose conversion and, thereby, the commercial viability of the process. There is no teaching of steam recovery.

[0014] U.S. Patent No. 5,628,830 (Brink) teaches a steam pre-treatment of biomass. The lignocellulosic material is finely ground, subjected to a first acid hydrolysis (pre-treatment) stage, then passed through a disintegrator. The slurry then undergoes one or more flashing stages to further reduce the particle size of the solids. The solids are then hydrolyzed with cellulase and the sugars are fermented to ethanol. There is no teaching of flash steam recovery.

[0015] U.S. Patent No. 4,237,226 (Grethlein) teaches a continuous pre-treatment in which cellulosic material is slurried in water to about 5-10% solids. The slurry is heated to the reaction temperature and a concentrated stream of sulfuric acid is injected. The acidified slurry is then heated by the injection of live steam to allow for very rapid heating of the slurry. Rapid

cooling to quench the reaction, by flashing across an orifice or capillary at the outlet to the reactor is described. However, recovery of steam is not described.

[0016] U.S. Patent No. 4,556,430 (Converse) includes a non-aqueous carrier in the feedstock to decrease the amount of water present. However, the carrier must be recovered. As some of the lignin may be dissolved by the carrier, recovery of the lignin and carrier is complex. Furthermore, there is no teaching of steam recovery.

[0017] EP 1,316,620 (Pinatti and Soares) disclose a batch hydrolysis process. The process involves feeding biomass with a helical screw feeder to a batch pre-hydrolysis reactor where the biomass is treated by the addition of steam and a pre-heated acidic solution. The acidic solution is pre-heated by heat exchange with pre-hydrolysate (sugar solution) that is withdrawn from the reactor and by escape steam that exits the reactor during the initial heating and degasifying operation. A limitation of the process is that heat is not recovered from the biomass solids and solution that remains in the reactor after the pre-hydrolysate solution is withdrawn. Furthermore, there is no teaching of carrying out a continuous pre-treatment process or of pumping the biomass feedstock.

[0018] U.S. Patent Nos. 6,419,788 and 6,620,292 (Wingerson) disclose a hydrolysis process for producing relatively pure cellulose from lignocellulosic materials. This first entails pre-heating mechanically disintegrated feedstock with steam. The pre-heated feedstock is then forced mechanically into a hydrothermal wash chamber where it is subjected to wet oxidation and washing with steam and alkaline wash water containing dissolved oxygen. The goal of the wet oxidation and washing step is to remove lignin, while preventing the hydrolysis of cellulose. Cellulose is discharged from the wash chamber into a flash tank and steam generated in the flash cooling is recycled to pre-heat the incoming feedstock. In order to further reduce the energy requirements of the system, the alkaline wash water is pre-heated by a heat recovery system that utilizes heat energy from discharged wash liquor. This heat energy is recovered by flashing the wash liquor in a chain of flash tanks and steam generated from each flash tank is introduced to a heat exchanger where it heats the incoming wash water. A disadvantage of Wingerson's process is that the recovery of steam from the single flash of cellulosic product to atmospheric pressure is inefficient. In addition, the feedstock is at a moisture content that is too low to allow it to be

pumped. Furthermore, the process requires separating the alkaline wash water used for wet oxidation from the hydrolyzed biomass under reactor pressure. In the processing of lignocellulosic material by acid pre-treatment, it is difficult and expensive to separate the acid pre-treated solids from liquor because of the muddy texture of such acidified slurries. Finally, the alkaline wash used by Wingerson is not applicable to an acid pre-treatment.

[0019] U.S. Patent No. 3,212,932 (Hess *et al.*) discloses a process for hydrolyzing lignocellulosic material to produce pentose and hexose sugars. The process involves pre-mixing finely divided lignocellulosic material with an aqueous solution (for example, water or water and acid) and introducing the mixture to a reaction vessel where it is heated by direct steam injection in a first stage hydrolysis. The material is subsequently subjected to a rapid reduction in pressure or "flash blowdown". The flash steam may be used for heat exchange with the starting material. The soluble pentose and hexose sugars are separated from the lignin and cellulose. The lignin and cellulose are treated with mineral acid and then subjected to a second stage of hydrolysis to convert the cellulose to hexose sugars under more strenuous conditions. This is carried out by heating the lignin and cellulose in a reactor with steam at a pressure of between 150 and 900 psig. After heating by steam, the treated material is subjected to a flash blowdown and the steam is vented. The solid product is washed with water and the liquor stream is then treated with a neutralizing agent and filtered to produce a product containing hexose. A disadvantage of the process of Hess *et al.* is that the recovery of steam in the first stage hydrolysis by a single flash is inefficient. In addition, there is no disclosure of pumping the lignocellulosic material through the system.

[0020] U.S. Patent No. 4,468,256 (Hinger) discloses a process for the hydrolysis of cellulose in vegetable raw materials. The raw material is comminuted and impregnated with dilute acid, after which excess moisture is removed from the material. The acidified material is pre-heated using recycled steam and is then fed into a reactor, where it is heated in one or more heating zones. High pressure steam is injected into the material in a hydrolysis zone. The material is subsequently cooled in one or more release zones, where pressure is rapidly released; the released steam is recycled to the heating zone. Residual steam remaining in the treated materials exits through a port and is recycled to the initial pre-heating stage. The feedstock of Hinger has a moisture content that is too low to allow pumping of the feedstock. Also, an

endless piston chain within the reactor is used to convey the feedstock into each zone of the reactor, which adds complexity and cost to the process. The piston chain is exposed to acid, which requires that it be constructed of acid resistant material, which adds considerable cost. Hinger also discloses a step of removing excess water after acid impregnation in order to keep the moisture content of the material as low as possible; however, dehydrating acidified feedstock upstream of the reactor is costly.

[0021] In the chemical pulping of wood chips, it is known to recover energy from consumed cooking chemicals remaining after cooking of the pulp. This may involve flashing of spent cooking liquor (black liquor) after separation from the chips in a series of flash tanks to produce a source of steam. This steam can be used in upstream processing steps, including steam pre-treatment of the wood chips in a chip bin. Pulping processes that make use of flashing steps to recover steam from black liquor are described in U.S. Patent Nos. 6,176,971, 6,179,958, 5,089,086, 6,346,166, 6,722,130 and 6,306,252. These patents do not disclose the processing of feedstock to convert cellulose to glucose. Furthermore, there is no teaching of producing a feedstock slurry that can be pumped through a pre-treatment reactor. The above patents also require separation of the cooked wood chips from the black liquor under digester pressure, with the liquor then being flashed. In the processing of lignocellulosic material by acid pretreatment, it is difficult and expensive to separate acid pre-treated material from liquor because of its muddy texture.

[0022] At present, methods of feedstock pre-treatment prior to enzymatic hydrolysis are not economic. The known methods for steam recovery are inefficient or may not be economic. Also, many methods require a high steam usage to heat the water of slurried feedstocks. There is a need in the art for a continuous pre-treatment process with significant steam recovery and that provides a feedstock that is pumpable.

SUMMARY OF THE INVENTION

[0023] The present invention relates to a pre-treatment process in the conversion of lignocellulosic feedstocks into sugars. More specifically, the present invention relates to a continuously flowing pre-treatment process with simultaneous steam recovery, for the conversion of lignocellulosic feedstocks into sugars.

[0024] It is an object of the present invention to provide a continuous flowing pre-treatment system with steam recovery. The pre-treatment system operates at reduced cost when compared to currently available methods.

[0025] The present invention provides a continuous process for pre-treating a lignocellulosic feedstock, comprising:

- (a) providing a pumpable feedstock slurry having a solids concentration of 8% to 30% by weight;
- (b) pumping the feedstock slurry through a heating train of two, or more than two stages, each heating stage including a pump to increase heating stage pressure and a direct steam injection to heat the feedstock slurry;
- (c) adding acid to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train, the acid added at a concentration of 0.2% to 12% weight of acid on weight of initial feedstock, to produce a heated, acidified feedstock slurry;
- (d) flowing the heated, acidified feedstock slurry through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes;
- (e) cooling the slurry after the slurry exits the pre-treatment reactor using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing; and
- (f) using steam energy from two or more than two of the flashing stages, at or near their flash pressures, to heat the feedstock slurry in the heating train.

[0026] The present invention also provides the process defined above, wherein the feedstock is agricultural wastes such as corn stover, soybean stover, corn cobs, rice straw, rice hulls, corn fiber, wheat straw, barley straw, canola straw, oat straw and oat hulls; grasses such as switch grass, miscanthus, cord grass, rye grass and reed canary grass; forestry biomass such as

recycled wood pulp fiber, softwood, hardwood and sawdust; sugar processing residues such as bagasse and beet pulp; or any combination thereof.

[0027] The present invention pertains to the process defined above, wherein the feedstock is slurried in an aqueous solution, such as water. The present invention is also directed to the method as just described, wherein the heating train comprises 2 to about 8 heating stages. The present invention is also directed to the method as just described, wherein live steam is added to one or more than one heating stage to heat the feedstock slurry. Furthermore, the step of cooling (step e), as just described, may comprise 2 to about 8 flashing stages. After the final flashing stage, the slurry may subsequently be cooled to a temperature of about 30°C to about 100°C.

[0028] The present invention pertains to the process described above, wherein the acid is sulfuric acid, sulfurous acid, sulfur dioxide, or any type of acid known in the art.

[0029] The present invention is also directed to the method as just described, wherein the acid is added to the feedstock slurry prior to the heating train. Alternatively, the acid may be added to the feedstock slurry after the heating train, or may be added to the feedstock slurry in a heating stage, for example, a final heating stage.

[0030] The present invention pertains to the process defined above, wherein the feedstock is reacted at a temperature of less than about 170°C in the presence of about 0.5% to about 3% weight of acid on weight of initial feedstock to hydrolyze the hemicellulose after the step of adding (step c) and prior to the step of flowing (step d).

[0031] The present invention is also directed to the method as described above, wherein the slurry is submitted to enzymatic hydrolysis subsequent to the step of cooling (step e).

[0032] The process of the present invention overcomes the disadvantages of the prior art by pumping a feedstock slurry into a continuous pre-treatment system and by using multiple heating stages corresponding to the pressures of the steam streams that are available. Steam at various pressures is obtained in the flashing stages, where it is captured and the steam or a portion of the steam is used, to heat the feedstock in the heating stages prior to the pre-treatment reactor. The process has the benefits of avoiding the inefficiencies associated with increasing

steam pressure in the flash train through the use of flash steam compressors. The process also avoids the inefficiencies associated with a single flash to lower temperature. Furthermore, the feedstock is slurried in liquid at a solids concentration of between 8% and 30% by weight solids, which is pumped through a heating train.

[0033] Therefore, the invention offers significant advances in the pre-treatment of lignocellulosic feedstock.

[0034] This summary of the invention does not necessarily describe all necessary features of the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

[0035] These and other features of the invention will become more apparent from the following description in which reference is made to the appended drawings wherein:

[0036] Figure 1 shows a schematic diagram of a non-limiting example of the process of a continuous flowing pre-treatment process according to the present invention.

DESCRIPTION OF PREFERRED EMBODIMENT

[0037] The present invention relates to a pre-treatment process in the conversion of lignocellulosic feedstocks into sugars. More specifically, the present invention relates to a continuously flowing pre-treatment process with simultaneous steam recovery, for the conversion of lignocellulosic feedstocks into sugars.

[0038] The following description is of a preferred embodiment by way of example only and without limitation to the combination of features necessary for carrying the invention into effect.

[0039] The present invention provides a continuous pre-treatment process of lignocellulosic feedstocks that involves pre-heating a pumpable feedstock slurry using two or more than two successive heating stages. Each stage is at a higher temperature than the previous stage. The higher temperatures are successively achieved by recycling the energy in the flash steam from the pre-treatment process. The final heat-up of the feedstock prior to the pre-

treatment reactor may be by injection of live steam. Acid is also added to the feedstock, and the heated, acidified feedstock is held in a pre-treatment reactor for a time sufficient to improve the efficiency of the hydrolysis of cellulose in the feedstock by cellulase enzymes. The pre-treated material is then subjected to two or more than two successive flashing stages to cool down the feedstock slurry and to remove the steam for re-use. There is no intermittent increase in steam temperature or pressure from the flashing stages. The flashed steam is captured and sent to the heating stages to be used with minimal loss of pressure, ensuring efficient recovery of the energy in the steam.

[0040] According to an embodiment of the present invention there is provided a continuous process for pre-treating a lignocellulosic feedstock, comprising:

- (a) providing a pumpable feedstock slurry having a solids concentration of 8% to 30% by weight;
- (b) pumping the feedstock slurry through a heating train of two, or more than two heating stages, each heating stage including a pump to increase stage pressure and a direct steam injection to heat the feedstock slurry;
- (c) adding acid to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train, the acid being added at a concentration of 0.2% to 12% weight of acid on weight of initial feedstock, to produce a heated, acidified feedstock slurry;
- (d) flowing the heated, acidified feedstock slurry through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes;
- (e) cooling the slurry after the slurry exits the pre-treatment reactor, using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing; and
- (f) using steam energy from two or more than two of the flashing stages, at or near their flash pressures, to heat the feedstock slurry in the heating train.

[0041] By "continuous process", it is meant a process that involves continuous feeding of feedstock and withdrawal of pre-treated feedstock.

[0042] By the term "lignocellulosic feedstock" or "lignocellulosic material", it is meant any type of biomass comprising cellulose such as, but not limited to, non-woody plant biomass, agricultural wastes, forestry biomass and sugar-processing residues. Generally, a lignocellulosic material is recognized as containing cellulose in an amount greater than about 20% (w/w), about 15% or more hemicellulose, and about 15% lignin. The cellulosic material can be of higher cellulose content, for example at least about 30% (w/w), 35% (w/w), 40% (w/w) or more. For example, the cellulosic material may comprise from about 20% to about 50%(w/w) cellulose, or more, or any amount therebetween, for example but not limited to 20, 22, 24, 26, 28, 30, 32, 34, 36, 38, 40, 42, 44, 46, 48, and 50% (w/w) cellulose.

[0043] In a non-limiting example, the lignocellulosic feedstock can include, but is not limited to grasses, such as switch grass, miscanthus, cord grass, rye grass and reed canary grass, or a combination thereof; sugar-processing residues such as, but not limited to sugar cane bagasse and beet pulp; agricultural wastes such as, but not limited to rice straw, rice hulls, corn cobs, barley straw, wheat straw, canola straw, oat straw, oat hulls, and corn fiber; stover, such as, but not limited to soybean stover and corn stover; and forestry biomass, such as, but not limited to recycled wood pulp fiber, sawdust, hardwood, softwood, or any combination thereof. Lignocellulosic feedstock may comprise one species of fiber, or alternatively lignocellulosic feedstock may comprise a mixture of fibers that originate from different lignocellulosic feedstocks. Agricultural wastes such as wheat straw, oat straw, barley straw, and canola straw; stovers such as corn stover and soybean stover; grasses such as switch grass, reed canary grass, cord grass, and miscanthus; or combinations thereof are particularly advantageous as lignocellulosic feedstocks, due to their widespread availability and low cost.

[0044] The present invention may be practiced with lignocellulosic feedstock that has been subjected to size reduction. Size reduction by any suitable mechanical action is acceptable, including cutting, grinding, milling, agitation, shredding, refining, compression/expansion, pressing or other types of mechanical action. Several mechanical size reduction methods are well known in the art.

[0045] Preferably, about 70% to about 100% of the feedstock particles are of a length of about 0.05 cm to about 2.5 cm. Alternatively, the feedstock size may be reduced to particles of size of about 20 to about 60 mesh, or any amount therebetween; for example the particle size may be about 20, 25, 30, 35, 40, 45, 50, 55, or 60 mesh.

[0046] The lignocellulosic material is slurried in liquid to a solids content of about 8% to about 30% (w/w) suspended (undissolved) solids, or any amount therebetween; for example, the feedstock slurry may be about 8, 10, 12, 15, 18, 20, 22, 25, 28, 30% (w/w) suspended solids, or any amount therebetween. The liquid may be any suitable liquid known in the art. For example, but without wishing to be limiting, the liquid may be an aqueous solution, an organic liquid that is miscible or immiscible in water, or a combination thereof.

[0047] In a non-limiting example, the feedstock is slurried in an aqueous solution which may be water. Preferably, the aqueous solution is heated prior to addition to the feedstock. The aqueous solution can be re-circulated from elsewhere in the process. The amount of aqueous solution added makes pumping of the feedstock possible, particularly with a medium consistency (MC) pump, or other pump suitable for handling the slurry. The optimum amount of aqueous solution added depends on the physical properties of the feedstock, and can be determined by methods well known to a person of skill in the art. Excess quantities of aqueous solution will result in additional heat requirements and reduce efficiency of the process, while quantities that are too small will be absorbed by the feedstock and impact pumpability.

[0048] The liquid used to slurry the feedstock may be at a temperature that provides a feedstock slurry temperature of about 30°C to about 110°C, or any temperature therebetween; for example, the feedstock slurry may be at about 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100, 105 or 110°C, or any temperature therebetween. The feedstock and the liquid can be combined in a soaking tank, a drop leg toward a tank, a pump standpipe, or by any other suitable means.

[0049] The slurried feedstock is then pumped to the heating train. By the term "heating train", it is meant two or more than two stages of steam heating of the slurried feedstock, each stage at successively higher temperatures. The number of stages in a heating train should be high enough to provide the ability to use steam at the different pressures that are available, but

low enough so the cost of pumps and the complexity is reasonable. In a non-limiting example, 2 to about 8 stages, or any amount therebetween, may be used; for example, a heating train may have 2, 3, 4, 5, 6, 7 or about 8 stages. In each stage, the steam may be directly injected into each heating stage using steam mixers, which will be familiar to those of skill in the art (see Pulp Bleaching: Principles and Practice, Reeve and Dence, p. 539-568, which is incorporated herein by reference). This method results in a rapid, uniform heating of the slurry, thereby minimizing degradation of the feedstock.

[0050] The final heating stage may involve the injection of live steam immediately prior to the pre-treatment reactor. The choice of temperature of the slurry entering the pre-treatment reactor will depend on the retention time, acid concentration, feedstock used and degree of treatment required; the pre-treatment temperature is generally between about 160°C and about 280°C, or any temperature therebetween. For example, the temperature may be about 160, 170, 175, 180, 185, 190, 195, 200, 205, 210, 215, 220, 225, 230, 235, 240, 245, 250, 255, 260, 265, 270, 275, or 280°C. It will be understood by those of skill in the art that the condensation of steam in the slurry decreases the solids concentration.

[0051] Acid is added to the feedstock slurry, either prior to the heating train, during a heating stage, or after leaving the heating train. For example, but without wishing to be limiting in any manner, the acid may be added in the final heating stage. In this case, the acid may be added immediately prior to, or immediately after the point of live steam injection. If the acid is added prior to entry into the heating train, the metallurgy of the heating train should be selected to handle acidic conditions. The acid is added in a manner that allows it to mix into the slurry. The acid may be added as concentrated acid or, prior to addition, it may be diluted with water or an aqueous solution.

[0052] Any suitable acid may be used in the process of the present invention. For example, sulfuric acid, sulfurous acid, sulfur dioxide, or combinations thereof can be used. In a specific non-limiting example, sulfuric acid, which is inexpensive, water soluble at high concentrations, stable, non-volatile, and capable of catalyzing the reactions desired in pre-treatment, is used. The amount of acid added may vary, but should be sufficient to achieve a final concentration of acid of about 0.2% to about 12% w/w, or any amount therebetween, of

feedstock solids. The resulting pH of the slurry should be within the range of about 0.8 to about 2.0 or any pH therebetween. For example, the pH of the slurry may be between about 0.8, 0.9, 1.0, 1.1, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, or 2.0.

[0053] The pre-treatment reactor preferably is designed to maintain a plug flow of the slurry and may be constructed of any suitable material resistant to corrosion and erosion at reaction conditions. For example, but without wishing to be limiting, the reactor may be constructed of zirconium, tantalum, niobium, ceramic tiles, or other such resistant materials. A valve at the exit of the reactor may be employed for maintaining the back pressure necessary for maintenance of the pressure at a desired level.

[0054] The retention time required in the pre-treatment reactor will vary depending on temperature, acid concentration, feedstock used, and the degree of treatment desired. For example, the slurry could be retained in the reactor for about 0.05 to about 10 minutes, or any time therebetween; for example, the retention time can be about 0.05, 0.1, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5, 5.5, 6, 6.5, 7, 7.5, 8, 8.5, 9, 9.5, or 10 minutes.

[0055] Upon exiting the pre-treatment reactor, the slurry is subjected to two or more than two flashing stages to cool the material, and to remove steam and volatiles from the system. For example, from 2 to about 8 successive flashing stages, or any amount therebetween, can be performed; for example, there may be 2, 3, 4, 5, 6, 7, or about 8 flashing stages. The multiple flashing stages generate flash steam at different pressures. This permits multiple uses of the steam, contributing to the overall process economics. In this manner, the energy of the steam is used multiple times, as the steam can be used directly or indirectly, by heating a process stream.

[0056] At least a portion of the flash steam from at least two of the flash vessels is sent to the heating train for heating the feedstock slurry prior to the pre-treatment reactor. Flash steam enters the heating train at a stage having a slurry pressure and temperature that (1) are low enough to accept and condense the steam, and (2) high enough to use the steam energy efficiently. This operation significantly reduces the consumption of live steam.

[0057] The provision of two or more flashing stages in the process results in a successive cooling and pressure decrease of the slurry. Preferably, the flash steam from at least one flashing

stage from which at least a portion of the flash steam is recycled to the heating train has a pressure that is about 20% to about 99.5% of the inlet slurry pressure to the flashing stage, or any value therebetween. More preferably, such flash stream has a pressure that is about 25% to about 99% of the slurry inlet pressure to the flashing stage, or more preferably still, a pressure that is about 35% to about 95% of the inlet slurry pressure to the flashing stage, or any value therebetween.

[0058] Alternatively, the flashed steam, or a portion thereof, may be used to heat a second stream of water, steam, or a combination thereof, for example in a re-boiler. This second stream may then be sent to the heating train. This also leads to an efficient use of the steam. Furthermore, the steam, or a portion of the steam, from one or more flash stages can be used as a source of steam in the plant. This is a particularly attractive option for the low pressure steam from the later flash stages. It should be appreciated that volatile impurities may be removed from flash steam prior to it being used in the heating train. This may be carried out, for example, in a re-boiler.

[0059] The slurry is cooled to a temperature suitable for downstream processing in the final flash vessel or in one or more heat exchangers. The pre-treated, cooled feedstock is subjected to enzymatic hydrolysis with cellulases. Optionally, washing, solids separation, or other processes are carried out prior to enzymatic hydrolysis. Prior to enzymatic hydrolysis, the pH of the slurry is generally adjusted to about 4.5 to about 5.5, or any pH therebetween, using a suitable alkaline solution; for example, the pH may be adjusted to about 4.5, 4.6, 4.7, 4.8, 4.9, 5.0, 5.1, 5.2, 5.3, 5.4, or 5.5. However, the pH of the slurry can be higher or lower than about 4.5 to 5.5 if the cellulase enzymes used are alkalophilic or acidophilic, respectively. It remains that the pH of the slurry should be adjusted to within the range of optimum pH for the enzymes used. For pH adjustments, any suitable alkaline solution known in the art can be used, for example, but not limited to sodium hydroxide, potassium hydroxide, ammonium hydroxide, ammonia, or calcium hydroxide.

[0060] For enzymatic hydrolysis, the optimum temperature is about 30°C to about 70°C, the temperature range depending on the cellulase enzymes used. Generally, a temperature in the range of about 45°C to about 55°C, or any temperature therebetween, is suitable for most

cellulase enzymes; for example, the temperature may be about 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, or 55°C. However, the temperature of the slurry may be higher for thermophilic cellulase enzymes. For example, but without wishing to be limiting, the slurry is cooled to about 50°C for hydrolysis performed with cellulase enzymes. A non-limiting example of a cellulase enzyme mixture may be made by the fungus *Trichoderma*.

[0061] By the term “cellulase enzymes”, “cellulase”, or “enzymes”, it is meant enzymes that catalyse the hydrolysis of cellulose to products such as glucose, cellobiose, and other cello-oligosaccharides. Cellulase is a generic term denoting a multienzyme mixture comprising exo-cellobiohydrolases (CBH), endoglucanases (EG) and β -glucosidases (β G) that can be produced by a number of plants and microorganisms. The process of the present invention can be carried out with any type of cellulase enzymes, regardless of their source. Among the most widely studied, characterized, and commercially produced cellulases are those obtained from fungi of the genera *Aspergillus*, *Humicola*, and *Trichoderma*, and from the bacteria of the genera *Bacillus* and *Thermobifida*. Cellulase produced by the filamentous fungi *Trichoderma longibrachiatum* comprises at least two cellobiohydrolase enzymes termed CBHI and CBHII and at least 4 EG enzymes.

[0062] Cellulase enzymes work synergistically to degrade cellulose to glucose. CBHI and CBHII generally act on the ends of the glucose polymers in cellulose microfibrils liberating cellobiose (Teeri and Koivula, 1995, Carbohydr. Europe 12, 28–33) while the endoglucanases act at random locations on the cellulose. Together these enzymes hydrolyse cellulose to smaller cello-oligosaccharides such as cellobiose. Cellobiose is hydrolysed to glucose by β -glucosidase.

[0063] The cellulase enzyme dosage added to the slurry is chosen to achieve a sufficiently high level of cellulose conversion. For example, an appropriate cellulase dosage can be about 5.0 to about 50.0 Filter Paper Units (FPU or IU) per gram of cellulose, or any amount therebetween. For example, the cellulase dosage may be about 5, 8, 10, 12, 15, 18, 20, 22, 25, 28, 30, 32, 35, 38, 40, 42, 45, 48, or 50 FPU, or any amount therebetween. The FPU is a standard measurement familiar to those skilled in the art and is defined and measured according to Ghose (1987, Pure and Appl.Chem. 59:257-268). An adequate quantity of β -glucosidase (cellobiase) activity is also added to the mixture. The dosage level of β -glucosidase may be

about 5 to about 400 β -glucosidase units per gram of cellulose, or any amount therebetween, or from about 35 to about 100 β -glucosidase units per gram of cellulose; for example, the dosage may be 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, 100, 125, 150, 175, 200, 225, 250, 275, 300, 325, 350, 375, or 400 β -glucosidase units per gram of cellulose, or any amount therebetween. The β -glucosidase unit is measured according to the method of Ghose (1987, Pure and Appl.Chem. 59:257-268).

[0064] The enzymatic hydrolysis continues for about 24 to about 250 hours, or any amount of time therebetween, depending on the degree of conversion desired. For example, the reaction time could be about 24, 30, 40, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140, or 150 hours, or any amount therebetween. The resulting slurry is an aqueous solution of glucose and xylose, and other carbohydrates, with lignin and other unconverted, suspended solids. The sugars are readily separated from the suspended solids and may be further processed as required, for example but not limited to fermentation to ethanol by yeast.

[0065] In another non-limiting example of the present invention, a two-stage pre-treatment system may be used. In the first stage, the acidified feedstock is heated in the heating train to a temperature below about 170°C. The feedstock is then reacted at about 170°C in the presence of about 0.5% to about 3% weight of acid on weight of initial feedstock to hydrolyze the hemicellulose prior to the main pre-treatment reaction. The solubilized hemicellulose can optionally be washed away from the slurry with water, thereby preventing degradation of the sugar in the second stage of pre-treatment. The remaining feedstock is then submitted to the second stage of pre-treatment involving the conditions outlined above. For example, the second stage of pre-treatment may involve pumping the feedstock slurry through a heating train of two, or more than two heating stages, each heating stage including a pump to increase stage pressure and a direct steam injection to heat the feedstock slurry. Following the addition of acid to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train, the heated, acidified feedstock slurry is passed through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase the efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes. After the slurry exits the pre-treatment reactor, the slurry is at least partially cooled using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing, and

the steam energy from two or more than two of the flashing stages, at or near its flash pressure is used to heat the feedstock slurry in the heating train.

[0066] With reference to Figure 1, which is a non-limiting example of the process, the feedstock **10** may be prepared by a shearing or crushing operation, or a combination thereof, resulting in small particles. The feedstock is then slurried in a drop leg **30**.

[0067] In the first heating stage **60** of the heating train, the feedstock slurry is pumped by pump **40** to steam mixer **65**. Pump **40** may be an MC® (medium consistency) pump to handle the slurry. Pump **40** may be connected to a first vacuum pump **55** that de-aerates the slurry. The heat to this stage is provided by direct injection of steam **60**.

[0068] Heated slurry **70** from the first heating stage is pumped by pump **80**, to the second stage steam mixer **105**. Flash steam **100** is injected into this stream to heat the stream.

[0069] The heated slurry **110** from the second stage **90** of the heating train is pumped by pump **120** to steam mixers **135** and **145**. The stream **130** is heated with live steam **140** added at two locations to create stream **150**. At this point, sulfuric acid **160** is added to the slurry to produce stream **170**. The acid may be mixed with mixing tee **165**. Stream **170** is the feed stream for the pre-treatment reactor **180**.

[0070] The pre-treatment reactor **180** may be of any suitable configuration, for example, a cylindrical pipe designed for a plug flow of the slurry. The inlet stream flows through the pre-treatment reactor **180** for a time sufficient to solubilize the hemicellulose and a smaller portion of the cellulose. Upon exiting the pre-treatment reactor **180**, the slurry **190** is flashed in a first flash tank **200**. The flash steam and volatiles stream **210** is used as the steam injection **100** in the second heating stage **90**.

[0071] The slurry **215** from the first flash tank **200** is flashed in a second flash tank **220**. The steam and volatiles stream **230** is used to heat, via line **60**, the first stage of the heating train **50**.

[0072] The slurry **240** from the second flash tank **220** is flashed in a third flash tank **250**. A portion of the volatile stream **260** is sent to a sparger tank **280** which pre-heats water **20**. The remaining portion **290** may be used as a source of low pressure steam **300** for the plant.

[0073] The slurry **310** from the third flash tank **250** is flashed in a fourth flash tank **320**. The steam and volatiles stream **330** is condensed at heat exchanger **340** and the condensate is added to the sparger tank **280** via line **350**. The heat exchanger fluid may be water stream **360** that flows out of the heat exchanger **340** and is used as boiler feed water **370**. Pump **380** is a vacuum pump that removes non-condensibles (**390**) from the flash stream.

[0074] The slurry stream **400** from the fourth flash tank is pumped by pump **410** to heat exchanger **420**. Upstream of the heat exchanger, ammonia **430** is injected into the slurry to adjust the pH up to about 4.5 to 5.5. The ammonia may be mixed with mixing tee **435**. Process water stream **25** is the heat exchange fluid. The tempered water is added to the sparger tank **280**, where it is heated by streams **350** and the condensate from the third flash tank **270**. The hot water **20** from the sparger tank **280** is used to slurry the feedstock in the initial drop leg **30**. The cooled slurry **440** at about 70°C may then be cooled to about 50°C and sent for enzymatic hydrolysis **450**.

[0075] The present invention will be further illustrated in the following examples. However, it is to be understood that these examples are for illustrative purposes only, and should not be used to limit the scope of the present invention in any manner.

Example 1

[0076] With reference to Figure 1, the feedstock **10**, for example, but not limited to, wheat straw may be prepared by a shearing or crushing operation, or a combination thereof, resulting in small particles at 54% moisture. About 70,000 kg/hr wheat straw (dry basis) is slurried in 370,000 kg/hr water **20** at a temperature of about 99°C in a drop leg **30** to reach a solids content of 13.4%.

[0077] In the first heating stage **50** of the heating train, the feedstock slurry is conveyed by pump **40** to steam mixer **65**. Pump **40** may be an MC® (medium consistency) pump to handle the slurry. Pump **40** may be connected to a first vacuum pump **55** that de-aerates the

slurry. The heat to this stage is provided by direct injection of 35,585 kg/hr steam **60** via steam mixer **65**, for example at about 166°C. This injection heats the slurry to about 124°C.

[0078] The first stage slurry **70** is conveyed by pump **80** to the second stage steam mixer **105**. Flash steam **100** at about 192°C is injected via steam mixer **105** at a rate of 40,670 kg/hr into this stream to heat the stream to about 162°C.

[0079] The second stage slurry **110** is conveyed by pump **120** to the third heating stage steam mixers **135** and **145**. The stream **130** is heated to about 193°C and then about 220°C with live steam at about 450 psig **140** added at about a rate of 38,939 kg/hr to each of the steam mixers **135** and **145**, respectively, to create stream **150**. Stream **150** from the third stage **115** of the heating train is at about 10% feedstock solids. At this point, sulfuric acid **160** is added to the slurry in an amount of about 4585 kg/hr to reach a concentration of about 6.55% acid by weight on initial feedstock solids to produce stream **170**. The acid is mixed with mixing tee **165**. Stream **170** is the feed stream for the pre-treatment reactor **180**.

[0080] The pre-treatment reactor **180** may be of any suitable configuration, for example, a cylindrical pipe designed for a plug flow of the slurry. The inlet stream flows through the pre-treatment reactor **180** in approximately two minutes. This is sufficient to solubilize about 95% of the hemicellulose, and about 10% of the cellulose, with about 12% degradation of the monomeric sugars. Upon exiting the pre-treatment reactor **180**, the slurry **190** is flashed from about 368 psig to about 175 psig in a first flash tank **200**. The flash steam and volatiles stream **210** is at a flow rate of about 40,670 kg/hr and is used as the steam injection **100** in the second heating stage **90**.

[0081] The slurry **215** from the first flash tank **200** is at a flow rate of about 641,744 kg/hr and is flashed from about 175 psig to about 88 psig in a second flash tank **220**. The steam and volatiles stream **230** is at a rate of about 35,585 kg/hr and is used to heat, via line **60**, the first stage of the heating train **50**.

[0082] The slurry **240** from the second flash tank **220** is at a rate of about 606,159 kg/hr and is flashed from about 88 psig to about 15 psig in a third flash tank **250**. About 18% of the volatile stream **260**, which is about 8950 kg/hr at about 121°C (**270**), is sent to a sparger tank **280**.

which pre-heats water 20. The remaining 82% (290) which is about 41,642 kg/hr may be used as a source of low pressure steam for the plant 300.

[0083] The slurry 310 from the third flash tank 250 which is about 555,567 kg/hr is flashed from about 15 psig to about 6 psig in a fourth flash tank 320. The steam and volatiles stream 330 is at a flow rate of about 11,551 kg/hr at about 110°C and is condensed at heat exchanger 340 and added to the sparger tank 280 via line 350. The heat exchanger fluid is water stream 360 that flows out of the heat exchanger 340 and is used as boiler feed water 370. Pump 380 is a vacuum pump that removes non-condensibles (390) from the flash stream.

[0084] The slurry stream 400 from the fourth flash tank is at a flow rate of about 544,016 kg/hr and a temperature of about 110°C and is pumped by pump 410 to heat exchanger 420. Prior to the heat exchanger, ammonia 430 is injected into the slurry to adjust the pH up to 4.5 to 5.5. The ammonia is mixed with mixing tee 435. Process water stream 25 is the heat exchange fluid. The tempered water is added to the sparger tank 280, where it is heated by streams 350 and the condensate from the third flash tank 270. The hot water 20 from the sparger tank 280 is used to slurry the feedstock in the initial drop leg 30. The cooled slurry 440 at about 76°C is then cooled to about 50°C and sent for enzymatic hydrolysis 450.

Example 2

[0085] This example illustrates the increase in the amount of live steam required to heat a feedstock slurry in a pre-treatment process which utilizes only a single flash stage to cool the slurry after the slurry exits the pre-treatment reactor. The flash steam from this flash stage is used for heating of the feedstock slurry in a heating train before it enters the pre-treatment reactor.

[0086] The live steam requirement for final heating of the slurry to the pre-treatment reactor temperature has been calculated on the basis of using the same conditions specified in Example 1, with the exception that only a single flash stage is used for cooling the pre-treated slurry and for generating flash steam.

[0087] Accordingly, about 70,000 kg/hr wheat straw (dry basis) which has been prepared to provide small particles of wheat straw at 54% moisture is slurried in 370,000 kg/h water at a temperature of about 99°C to reach a solids content of 13.4%.

[0088] The feedstock slurry is pumped to one or more steam mixers in which flash steam at 110°C is used to heat the slurry to about 109°C. The slurry is then heated to 220°C with live steam at 450 psig by direct injection of the steam into one or more steam mixers.

[0089] Sulfuric acid is added to the slurry in an amount to reach a concentration of 6.55% acid by weight on initial feedstock solids. The slurry enters the pre-treatment reactor and flows through the pre-treatment reactor in approximately two minutes. Upon exiting the pre-treatment reactor, the slurry is flashed from about 368 psig to 6 psig in the flash tank. The flash steam and volatiles from the flash tank are used in the heating train to heat the slurry. Any excess steam is used as a source of low pressure steam for the plant.

[0090] Under the above conditions using a single flash, 126,150 kg/h of live steam would be required to heat the slurry from 109°C to the pre-treatment reaction temperature of 220°C. By contrast, in the embodiment of the invention described in Example 1, the live steam requirement is only 77,878 kg/h. Thus, by way of comparison with Example 1, the live steam consumption increases by 62% when only a single flash is used.

[0091] The above description is not intended to limit the claimed invention in any manner. Furthermore, the discussed combination of features might not be absolutely necessary for the inventive solution.

THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A continuous process for pre-treating a lignocellulosic feedstock, comprising:
 - (a) providing a pumpable feedstock slurry having a solids concentration of 8% to 30% by weight;
 - (b) pumping the feedstock slurry through a heating train of two, or more than two heating stages, each heating stage including a pump to increase heating stage pressure and a direct steam injection to heat the feedstock slurry;
 - (c) adding acid to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train, the acid added at a concentration of 0.2% to 12% weight of acid on weight of initial feedstock, to produce a heated, acidified feedstock slurry;
 - (d) flowing the heated, acidified feedstock slurry through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes;
 - (e) cooling the slurry after the slurry exits the pre-treatment reactor, using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing; and
 - (f) using steam energy from two or more than two of the flashing stages, at or near their flash pressures, to heat the feedstock slurry in the heating train.
2. The process of claim 1, wherein, in the step of providing (step a), the feedstock is selected from the group consisting of agricultural waste, grass, forestry biomass, sugar processing residues and combinations thereof.
3. The process of claim 2, wherein the agricultural waste is selected from the group consisting of corn stover, soybean stover, corn cobs, rice straw, rice hulls, corn fiber, wheat straw, barley straw, canola straw, oat straw, oat hulls and combinations thereof.

4. The process of claim 2, wherein the grass is selected from the group consisting of switch grass, miscanthus, cord grass, rye grass, reed canary grass and combinations thereof.
5. The process of claim 2, wherein the forestry biomass is selected from the group consisting of recycled wood pulp fiber, softwood, hardwood, sawdust and combinations thereof.
6. The process of claim 2, wherein the sugar processing residue is bagasse, beet pulp or combinations thereof.
7. The process of claim 1, wherein, in the step of providing (step a), the feedstock is slurried in an aqueous solution.
8. The process of claim 7, wherein, in the step of pumping (step b), the heating train comprises 2 to 8 heating stages.
9. The process of claim 8, wherein the step of cooling (step e) comprises 2 to 8 flashing stages.
10. The process of claim 9, wherein the slurry is subsequently cooled to about 30°C to about 100°C after the final flashing stage.
11. The process of claim 1, wherein, in the step of adding (step c), the acid is selected from the group consisting of sulfuric acid, sulfurous acid, and sulfur dioxide.
12. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry prior to the heating train.
13. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry after the heating train.
14. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry in a heating stage.
15. The process of claim 1, wherein, after the step of adding (step c) and before the step of pumping (step d), the feedstock slurry is reacted at a temperature of less than about

170°C in the presence of about 0.5% to about 3% weight of acid on weight of initial feedstock to hydrolyze the hemicellulose.

16. The process of claim 1, wherein, after the step of cooling (step e), the slurry is submitted to enzymatic hydrolysis by cellulase enzymes.
17. The process of claim 1, wherein, in the step of pumping (step b), live steam is added to one or more than one heating stage to heat the feedstock slurry.

AMENDED CLAIMS

received by the International Bureau on 08 February 2006 (08.02.2006). Original claims 1 and 18 have been replaced by amended claims 1-18 (3 pages).

THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A continuous process for pre-treating a lignocellulosic feedstock, comprising:
 - (a) providing a pumpable feedstock slurry having a solids concentration of 8% to 30% by weight;
 - (b) pumping the feedstock slurry through a heating train of two, or more than two heating stages, each heating stage including a pump to increase heating stage pressure and a direct steam injection to heat the feedstock slurry;
 - (c) adding acid to the feedstock slurry prior to the heating train, during a heating stage, or after leaving the heating train, the acid added at a concentration of 0.2% to 12% weight of acid on weight of initial feedstock, to produce a heated, acidified feedstock slurry;
 - (d) flowing the heated, acidified feedstock slurry through a pre-treatment reactor at a temperature of 160°C to 280°C for a time sufficient to increase efficiency of conversion of cellulose in the feedstock to glucose using cellulase enzymes;
 - (e) cooling the slurry after the slurry exits the pre-treatment reactor, using two, or more than two flashing stages at successively lower pressures, without increasing the steam pressure after flashing; and
 - (f) using at least a portion of the steam energy from two or more than two of the flashing stages to heat the feedstock slurry in the heating train, wherein flash steam from at least one of the flashing stages, from which at least a portion of the flash steam is used in the heating train, has a pressure that is 20% to 99.5% of the inlet slurry pressure to such flashing stage.

2. The process of claim 1, wherein, in the step of providing (step a), the feedstock is selected from the group consisting of agricultural waste, grass, forestry biomass, sugar processing residues and combinations thereof.
3. The process of claim 2, wherein the agricultural waste is selected from the group consisting of corn stover, soybean stover, corn cobs, rice straw, rice hulls, corn fiber, wheat straw, barley straw, canola straw, oat straw, oat hulls and combinations thereof.
4. The process of claim 2, wherein the grass is selected from the group consisting of switch grass, miscanthus, cord grass, rye grass, reed canary grass and combinations thereof.
5. The process of claim 2, wherein the forestry biomass is selected from the group consisting of recycled wood pulp fiber, softwood, hardwood, sawdust and combinations thereof.
6. The process of claim 2, wherein the sugar processing residue is bagasse, beet pulp or combinations thereof.
7. The process of claim 1, wherein, in the step of providing (step a), the feedstock is slurried in an aqueous solution.
8. The process of claim 7, wherein, in the step of pumping (step b), the heating train comprises 2 to 8 heating stages.
9. The process of claim 8, wherein the step of cooling (step e) comprises 2 to 8 flashing stages.
10. The process of claim 9, wherein the slurry is subsequently cooled to about 30°C to about 100°C after the final flashing stage.
11. The process of claim 1, wherein, in the step of adding (step c), the acid is selected from the group consisting of sulfuric acid, sulfurous acid, and sulfur dioxide.

12. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry prior to the heating train.
13. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry after the heating train.
14. The process of claim 1, wherein, in the step of adding (step c), the acid is added to the feedstock slurry in a heating stage.
15. The process of claim 1, wherein, after the step of adding (step c) and before the step of pumping (step d), the feedstock slurry is reacted at a temperature of less than about 170°C in the presence of about 0.5% to about 3% weight of acid on weight of initial feedstock to hydrolyze the hemicellulose.
16. The process of claim 1, wherein, after the step of cooling (step e), the slurry is submitted to enzymatic hydrolysis by cellulase enzymes.
17. The process of claim 1, wherein, in the step of pumping (step b), live steam is added to one or more than one heating stage to heat the feedstock slurry.
18. The process of claim 1, wherein, in the step of using steam energy (step f), flash steam, or a portion thereof, from at least one of the two or more flashing stages is used to heat a stream of water, steam, or a combination thereof, and wherein at least a portion of the heated stream is sent to the heating train to heat the feedstock slurry.

1/1

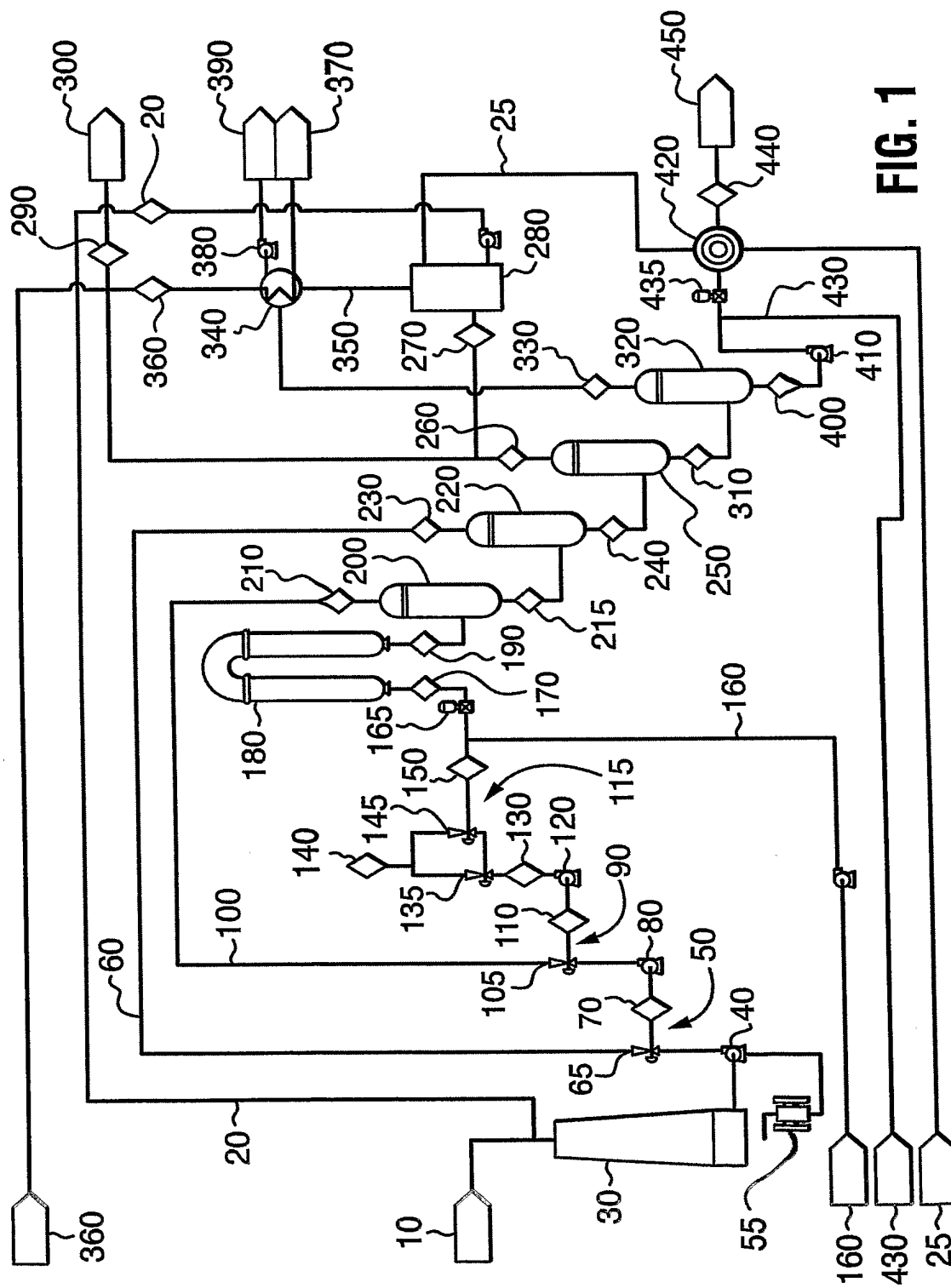


FIG. 1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CA2005/001493

A. CLASSIFICATION OF SUBJECT MATTER
IPC(7): B09B 3/00, C12P 7/10, C12P 19/02, C12P 19/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC(7) B09B 3/00, C12P 7/10, C12P 19/02, C12P 19/14, C12S 03/04.

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database(s) consulted during the international search (name of database(s) and, where practicable, search terms used)

Espacenet, USPTO, CPD, internet; keywords: feedstock, pump/s/ed/ing/able, acid/s/idified, glucose, flash/in_g, pressure, steam, process, method (singly or in combination, with/without class as appropriate).

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2,108,567 A (SCHOLLER, H., et al.) 15 February 1938 (15-02-1938)	1-17
A	US 2,681,871 A (WALLACE, L.C.) 22 June 1954 (22-06-1954)	1-17
A	US 3,212,932 A (HESS, R.W., et al.) 19 October 1965 (19-10-1965) *cited by the applicant*	1-17
A	US 4,137,094 A (HUGHES, J.F.) 30 January 1979 (30-01-1979)	1-17
A	US 4,237,226 A (GRETHLEIN, H.E.) 02 December 1980 (02-12-1980) *cited by the applicant*	1-17
A	US 4,316,747 A (RUGG, B.A., et al.) 23 February 1982 (23-02-1982)	1-17
A	US 4,318,748 A (CHURCH, J.A.) 09 March 1982 (09-03-1982)	1-17
A	US 4,427,453 A (REITTER, F.J.) 24 January 1984 (24-01-1984)	1-17
A	US 4,615,742 A (WRIGHT, J.D.) 07 October 1986 (07-10-1986)	1-17
A	US 6,620,292 B2 (WINGERSON, R.C.) 16 September 2003 (16-09-2003) *cited by the applicant*	1-17

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :
 "A" document defining the general state of the art which is not considered to be of particular relevance
 "E" earlier application or patent but published on or after the international filing date
 "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
 "O" document referring to an oral disclosure, use, exhibition or other means
 "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
 "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
 "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
 "&" document member of the same patent family

Date of the actual completion of the international search

07 November 2005 (07-11-2005)

Date of mailing of the international search report

7 December 2005 (07-12-2005)

Name and mailing address of the ISA/CA
 Canadian Intellectual Property Office
 Place du Portage I, C114 - 1st Floor, Box PCT
 50 Victoria Street
 Gatineau, Quebec K1A 0C9
 Facsimile No.: 001(819)953-2476

Authorized officer

Edward Dabrowski (819) 953-1378

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.
PCT/CA2005/001493

Patent Document Cited in Search Report	Publication Date	Patent Family Member(s)	Publication Date
US2108567 A	15-02-1938	NONE	
US2681871 A	22-06-1954	NONE	
US3212932 A	19-10-1965	NONE	
US4137094 A	30-01-1979	NONE	
US4237226 A	02-12-1980	NONE	
US4316747 A	23-02-1982	AT15074T T AU548717 B2 AU6812681 A BR8101561 A CA1190923 A1 DE3171880D D1 EP0037912 A2 IN153348 A1 JP1039760B B US4316748 A US4363671 A US4368079 A US4390375 A US4591386 A ZA8101415 A	15-09-1985 02-01-1986 24-09-1981 22-09-1981 23-07-1985 26-09-1985 21-10-1981 07-07-1984 23-08-1989 23-02-1982 14-12-1982 11-01-1983 28-06-1983 27-05-1986 30-06-1982
US4318748 A	09-03-1982	CA1152500 A1	23-08-1983
US4427453 A	24-01-1984	AT8275T T AU6789481 A BR8107083 A DE3164501D D1 EP0035679 A1 ES499625D D0 FI810523 A JP57500091T T NZ196322 A WO8102428 A1	15-07-1984 11-09-1981 05-01-1982 09-08-1984 16-09-1981 16-12-1981 24-08-1981 21-01-1982 16-12-1983 03-09-1981
US4615742 A	07-10-1986	NONE	
US6620292 B2	16-09-2003	AU8542401 A BR0113278 A CA2418726 A1 EP1328679 A1 US6419788 B1 WO0214598 A1	25-02-2002 08-07-2003 21-02-2002 23-07-2003 16-07-2002 21-02-2002